

Interactive comment on “The GESAMP atmospheric iron deposition model intercomparison study” by Stelios Myriokefalitakis et al.

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We thank the Referee#2 for the careful reading of the manuscript. Please find below the point-by-point answers to the referee's general, specific and technical comments.

1. General comments:

Q: My main concern is about the interannual variability of the models. As stated in Table 1, the simulated years are different for each model but the interannual variability of each model is not presented nor discussed.

A: This work uses single year model results only. Therefore, no year-to-year variability

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is possible to be presented in our post-processed analysis for the present atmosphere. However, our analysis shows the variability derived from the structural differences between models.

Q: Moreover, no requests for meteorological conditions or emission inventories have been set to the model simulations and the sensitivity of each model to these parameters are also not discussed.

A: The aim of this work is to describe the current state of Fe global atmospheric deposition modeling and provide a multi-model ensemble of Fe atmospheric deposition fluxes of the current estimates, characterized with regard to observations (see introduction). It aims also to understand the origin of the respective model differences over the oceanic regions among the participating models and not to conclude which model is the best. As discussed in the model description (section 2), the participating models use different parameterizations to simulate the Fe-cycle in the atmosphere. Although using the same meteorology and the same emission inventories in the participating models would have been a very interesting exercise this is out of the scope of the present study. All models are driven by publicly available meteorological datasets that have been used and evaluated in numerous studies. As shown in Table 1, two models are using GEOS-5, one model GEOS-FP and one ERA interim meteorology (see in Table 1).

2. Specific comments:

Q: P2, line 1: please add min and max for TFe and LFe deposition fluxes.

A: The minimum and maximum values for the global mean of the deposition fluxes TFe and LFe are added. This part now reads: “The mean global deposition fluxes into the global ocean is here estimated in the range of 10-30 Tg-Fe yr⁻¹ and 0.2-0.4 Tg-Fe yr⁻¹ for TFe and LFe, corresponding to roughly ~15 Tg-Fe yr⁻¹ and ~0.3 Tg-Fe yr⁻¹, respectively, for the for the multi model ensemble model mean.”

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Q: P3, lines 18-22: the fraction of Fe that is bioavailable is still not well known and also depends on phytoplankton species, so I suggest that the authors do not write that labile Fe is a good approximation for bioavailable Fe.

A: We agree with the reviewer that Fe bioavailability is a complex issue and for this we clearly stated this in the manuscript (pp3, lines: 16-22). However to make it more clear we rephrased this part as (please see also our reply to Reviewer 1): “The bioavailability of Fe is a complex issue (e.g., Lis et al., 2015; Morel et al., 2008) and several naming conventions and abbreviations were used to characterise the atmospheric supply of potentially bioavailable Fe to the global ocean (Baker and Croot, 2010; Shi et al., 2012). It has been widely assumed that soluble Fe can be considered, as a first approximation, to be bioavailable (Baker et al., 2006a, 2006b) and a common experimental practice to determine the bioavailable Fe fraction in Fe-containing aerosols is the quantification of Fe in a leachate solution that passes through 0.45 μm , 0.2 μm or 0.02 μm sized filter (see Meskhidze et al., 2016 and ref. therein). However, due to its operational definition, it has been shown that this filterable Fe may contain both the soluble Fe and colloidal forms (Jickells and Spokes, 2001; Raiswell and Canfield, 2012). Upon deposition to the surface ocean, the soluble of Fe delivered through atmospheric pathways can either enter the dissolved Fe pool in the ocean, or precipitate-out as large oxy-hydroxide particles (de Baar and de Jong, 2001; Boyd and Ellwood, 2010; Meskhidze et al., 2017; Turner and Hunter, 2001). Consequently, the impact of atmospheric Fe on marine biogeochemistry depends on both the total Fe (TFe) deposition and its solubility, keeping in mind that the bioavailable fraction of Fe in seawater will then also change due to post-atmospheric deposition ocean processes (e.g., Baker and Croot, 2010; Chen and Siefert, 2004; Meskhidze et al., 2017; Rich and Morel, 1990).”

Q: P4, line 8: “can be also be”

A: Corrected.

Q: P6, lines 2-5: for the role of oxalate on Fe solubility, Paris et al. (2011) could be

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cited as well (<https://doi.org/10.1016/j.atmosenv.2011.08.068>).

A: Reference added.

Q: P7, line 29: please change “in (Albani et al., 2014)” by “in Albani et al. (2014)”

A: Typo corrected.

Q: P18, lines 28-30: “LFe sources are mainly driven by mineral dust aerosols, although a significant fraction (6 to 62%) is due to LFe combustion aerosols, especially over the high-latitudes of the Northern Hemisphere (Ito et al. 2018; companion manuscript to be submitted).” I would rather put this sentence in the previous section.

A: We agree with the reviewer. The sentence has been moved into the previous section.

Q: P19, lines 1-5: the authors compare the seasonal variability of LFe, but I would have liked to see the error bars on Fig. 1, as well as more information on the statistical test (which one was used, P value, n, . . .).

A: Error bars in Fig. 1 are added. For the individual models, however, the results here correspond to one year of simulation. Therefore, no statistics can be derived for the seasonal deposition fluxes which are calculated as the sum of monthly deposition fluxes. We provide further statistics for the ensemble model; the median bias correction factors are presented in Table 3 for each model, together with the lower and upper 95% confidence interval.

Q: Moreover, the authors state that “in most of the cases IMPACT and GEOS-Chem present similar seasonal variation.” However, IMPACT is higher in JJA, while GEOS is higher in MAM.

A: We thank the reviewer for pointing this inconsistency. We now corrected this part as: “However, significant differences in the magnitude of the deposition fluxes are calculated between models (Fig. 1). A seasonal maximum in the deposition fluxes is calculated by CAM4 and GEOS-Chem during MAM, attributed to Saharan mineral dust

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aerosols, while IMPACT and TM4-ECPL present a seasonal maximum during JJA.”

Q: P19, lines 9-11: the authors state that “in the other seasons the 30N maximum is not clearly present”, but in JJA, a clear maximum for IMPACT is seen at 30°N.

A: We now rephrased this part as: “In DJF, and to a lesser extent in JJA, two zonal maxima are shown near the equator and around 30N.”

Q: P21, line 12: the authors should explain how they calculate the mean normalized bias. Why would a value of 2.4 mean that the concentrations are underestimated? This is not clear to me.

A: We agree with the reviewer that this sentence is confusing. We have rephrased for clarity as follows: “This reflects that overall the models overestimate TFe surface mass concentrations. However, from Fig. 4 we can see that this overestimate is higher for the highest TFe concentrations near the dust source regions and tend to turn to an underestimate for the lowest concentrations observed over remote oceans.” A detailed description of MNB calculations is also added in the manuscript (please see also our reply to Referee#1).

Q: P22, line 3 and Fig. 5: the authors discuss the relationship between Fe solubility and aerosol Fe concentrations but these 2 variables are not independent as the latter one is used to calculate the former one. How do the authors deal with that?

A: This is a good question. We are aware that concentrations and solubility are not independent variables in the calculations since solubility is the ratio of Labile to Total Fe concentrations. At low Fe concentration, for example, Fe solubility of TM4 is similar to ensemble model (red triangles in Fig. 5). This is because the mean Fe solubility is weighted by Fe concentration. More specifically, Fe concentration at low concentration of TM4 is much higher than other models, resulting in similar Fe solubility between TM4 and ensemble model. This indicates that a small number of aerosols with high Fe concentration can determine Fe solubility in bulk samples. We added further work

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to address one of the major quantification problems due to the sampling issues in p.25, l.2, “due to short of the sampling frequencies. A comparison of long-term measurements with multi-year hindcast will allow us to assess the model performance to simulate labile Fe concentration under specific events” after “Note, however, that evaluation of monthly mean model results by comparison with the shorter-term (e.g., daily) observations during different sampling periods introduces uncertainties”. At the same time, models consider the process of enhancement of Fe solubility. It is therefore interesting to see whether the models are able to capture the fraction of LFe to TFe correctly and this is what we are evaluating in Fig. 5. This figure shows that the models have difficulties to simulate the 4 orders of magnitude variability from 0.02% to 98% in the Fe solubility observed in the atmosphere (Fig. 5a). IMPACT simulates almost 3 orders of magnitude variability in Fe solubility. In the other models including the ensemble model, Fe solubility is less variable (one to two orders of magnitude only). In particular, low solubilities (high concentrations near sources) are overestimated and high solubilities (low concentrations at remote locations) are underestimated. This may indicate that the primary LFe in the models is overestimated and that models are missing solubilisation processes during transport or that those considered in the models are not sufficient effective. The discussion has been added appropriately in the manuscript.

Q: P23, line 12-: How is the lifetime (turnover time) calculated? Is it calculated by dividing the concentration by the deposition flux, both estimated by the model? This could be added in the text.

A: We explain in the caption of Fig. 6 that lifetimes are the calculated atmospheric concentrations (or burdens) divided by total sinks”, but for clarity we also added the following explanation in the manuscript: “Figure 6 presents the spatial distribution of TFe lifetime over the ocean (i.e., atmospheric concentrations divided by total sinks), as calculated for the ensemble model.”

Q: P24, lines 26-27: please change “similar to what it was pointed out in (Albani et al., 2014) and seen in the dust model intercomparison study of Huneus et al. (2011).”

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By “similar to what was pointed out in Albani et al. (2014) and seen in the dust model intercomparison study of Huneeus et al. (2011)”.

A: We corrected the typo: “(Albani et al., 2014)” to “Albani et al. (2014)”

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